Nonradiative Recombination Processes in GaN-based Semiconductors Probed by the Transient Grating Method

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Abstract

GaN-based semiconductors are very advantageous materials for LED and LD within blue-UV spectral region. Until now, a lot of studies have been done on the optical properties of GaN-based semiconductors by measuring the photoluminescence (PL), electricluminescence (EL), or cathodoluminescence (CL). Such measurements are based on the observation of radiative recombination processes of carriers and excitons in materials. On the other hand, direct observations of nonradiative recombination dynamics have not been attempted so far. However, both radiative and nonradiative processes should be known to elucidate the carrier dynamics and to develop the device properties because the internal quantum efficiency (η_{int}) is determined from the radiative and nonradiative recombination lifetime. In this paper, we observed the nonradiative recombination processes of GaN-based semiconductors directly by the transient grating (TG) method. The TG method is one of the third order nonlinear spectroscopic technique.[1] By using this method, the thermal dynamics of nonradiative recombination processes can be detected directly.[2]

Configuration of the excitation and probe beams of the TG method was shown in figure 1. The frequency-tripled beam of Nd:YAG laser (355nm) was used for the excitation beam. The interference pattern is created by crossing two excitation beams in sample materials. The nonradiative recombination process releases the thermal energy and the temperature of the sample is modulated (thermal grating). Optical properties of the materials such as refractive index and absorbance are also modulated by the photo excitation. Such modulation of optical properties results in the refractive grating. A probe beam from a He-Ne laser (633nm) was partly diffracted (TG signal) by these gratings. The whole measurements have been done at room temperature (23^{o}C).

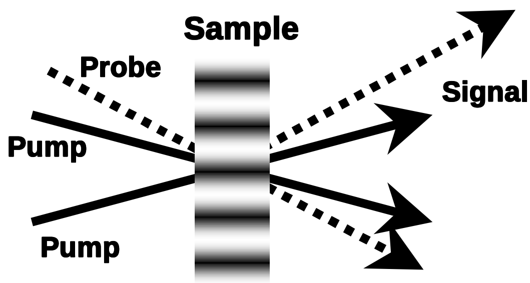

Figure 1 Configuration of the excitation and probe beams of the Transient grating (TG) method.

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The sample of 4μm-GaN was grown on sapphire (0001) by metalorganic chemical vapor deposition (MOCVD). For comparison, we used 0.9μm-ZnSe grown on ZnSe substrate by molecular beam epitaxisy (MBE).

Figure 2 shows the time profile of the TG signal taken for the GaN semiconductors. The signal rises immediately within the excitation pulse (few nanosecond). This decay profile can be fitted by a single exponential function. The diffraction efficiency of the TG signal is proportional to the square of refractive index change due to the increase of temperature (ΔT) originating from the nonradiative recombination. Therefore, the pre-exponential factor and rate constant obtained form fitting suggest the ΔT value by the nonradiative recombination and the thermal diffusivity (D_{th}), respectively. D_{th} is given by the obtained rate constant as 0.41 cm^2s^{-1} for GaN. This D_{th} value of GaN is about 5 times larger than that of ZnSe (0.084 cm^2s^{-1}) [2], indicating that GaN has merit for driving the heat out of an active layer. D_{th} can be calculated theoretically by the density (ρ), heat capacity (C_p), and thermal conductivity (λ_c) as $D_{th}=\lambda_c/\rho C_p$. By using the literature values (λ_c=1.3 $Wcm^{-1}K^{-1}$, ρ=6.095 gcm^{-3}, and C_p=9.745 $calmol^{-1}K^{-1}$), the theoretical value was calculated as D_{th}=0.44 cm^2s^{-1} which is close to the experimental value. This fact supports that the obtained TG signals were generated from the effect of thermal grating. The root square of the signal intensities is proportional to ΔT. We compared ΔT of GaN with that of ZnSe [2] under the several excitation energy densities (I_{ex}) as shown in figure 3. Dotted lines are guide for eyes. In the case of ZnSe, ΔT was saturated under the high I_{ex}. This fact suggests that the nonradiative recombination centers of ZnSe saturate at the high density of carriers or excitons. On the other hand, it was found that the nonratiative center in GaN was not saturated. In many semiconductors including ZnSe, dislocations of the crystals act as a nonradiative centers. However, recently, we reported that the nonradiative recombination lifetime of GaN is not so sensitive to the dislocation densities.[3] Such difference in the nature of nonradiative centers may reflect the variance of ΔT - I_{ex} characteristics between GaN and ZnSe. Similar measurement of InGaN/GaN is in progress and would give valuable information on the optical properties.

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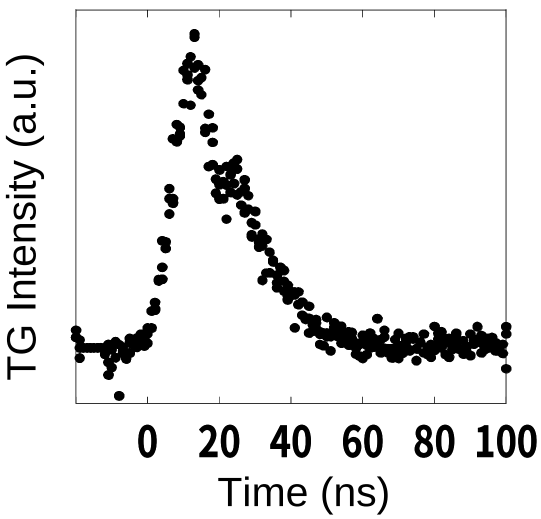

Figure 2 Time profile of the TG signal of the GaN epitaxial layer at room temperature.

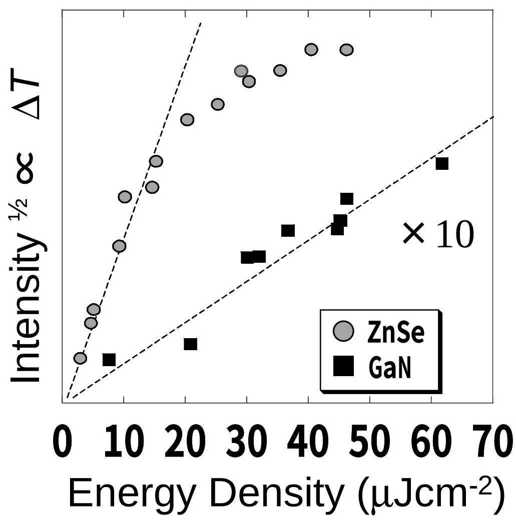

Figure 3 Excitation energy dependence of the root square of the TG signal of ●ZnSe and ■GaN

[1] H. J. Eichler, P. Gunter, and D. W. Pohl, Laser-Induced Dynamic Grating (Springer, Berlin, 1986)

[2] K. Okamoto, Y. Kawakami, Sg. Fujita and M. Terazima, *submitted for publication*.

[3] T. Izumi, Y. Narukawa, K. Okamoto, Y. Kawakami, Sg. Fujita and S. Nakamura, J. Lumin. 87-89 (2000) 1196.